British High Flux Beam Reactor

by
P. A. EGELSTAFF*
Atomic Energy Research Establishment, Harwell

The neutron scattering technique has become an accepted method for the study of condensed matter. Because of the great scientific and technical value of neutron experiments and the growing body of users, several proposals have been made during the past decade for a nuclear reactor devoted primarily to this technique. This article reviews the reasons for and history behind these proposals.

SLOW neutrons have been used widely in condensed matter physics, for many problems in chemistry and for industrial experiments. They represent a powerful tool of considerable versatility for several reasons. First, neutron wavelengths are about equal to the spacing between atoms in condensed matter while the corresponding energies are about equal to the spacing of the energy levels that determine chemical and physical properties of the material. The scattering by an isolated nucleus is isotropic and elastic so that there are no complications due to form factors, selection rules or competing cross-sections. Diffraction effects from neutrons scattered by nearby nuclei will show up atomic structure, and energy exchange between the neutron and matter gives a new determination of energy levels; simultaneous analysis provides new information.

Second, neutrons penetrate matter in bulk, having mean free paths of about 1 cm, so that surface effects can be neglected and a sample can be measured inside a furnace, cryostat or pressure container. Third, because neutron cross-sections are distributed virtually at random, both throughout the periodic table and between isotopes, light elements can be studied in the presence of heavy elements and the scattering properties of a sample can be changed by changing the isotopes without altering the chemistry of the sample. In addition the scattering of differently oriented but otherwise identical nuclei is different: for example, hydrogen with its two possible spin orientations scatters with a weak mean scattering amplitude and a strong random value. The incoherent scattering arising from such random effects contains no structural information, but makes possible the study of essentially "one-atom" dynamical effects.

Fourth, neutrons possess a magnetic moment so that magnetic atoms scatter neutrons through an effect which allows the structure and the dynamics of the electron spins, and hence of certain chemical bonds, to be studied. Finally, the range of wavelengths available extends from 0·3 to 10 Å and (by varying scattering angles) spacings or particle sizes between 0·1 Å and 20,000 Å can be measured.

This range of useful properties led to a wide usage of the technique once satisfactory neutron spectrometers became available. In many experiments it is possible to measure the strength, position and width of some peak in the dependence of scattering intensity on angle of scatter or on energy change of the neutron. Where such a well defined peak exists the interpretation is usually unique and quantitative in an absolute way. Experiments of this type are listed here as i–v. Some of the most exciting new applications, such as vi–x, however, arise where the scattering is not organized into well defined peaks, for example, the study of liquids, amorphous substances and disordered crystals. It is possible often to prepare samples that differ only in one feature, and a comparison of these samples allows the scattering by that feature to be studied, as in experiments xi and xii. Clearly neutron scattering experiments have an unparalleled richness, variety and depth, and touch on most of the problems connected with the several forms of condensed matter. Although neutron fluxes are low by comparison with, for example, X-ray or light fluxes the experiments are powerful. It is to extend the range and power of these experiments that the building of a high flux beam reactor (HFBR) is being considered. Extensive descriptions of the results of neutron experiments have been published[1,2], and I shall describe some of them briefly.

* Present address: Department of Physics, College of Physical Science, University of Guelph, Guelph, Ontario, Canada.

EXPERIMENTS IN WHICH NEUTRON SCATTERING HAS PROVED USEFUL

(i) Atomic structure of crystals deduced from Bragg reflexions in a way analogous to X-ray work.
(ii) Magnetization distribution in crystals deduced similarly; no alternative technique is available.
(iii) Frequency, wavelength, amplitude and polarization of crystal thermal vibrations, deduced from energy and momentum changes of neutrons; no alternative technique is available.
(iv) Thermally excited oscillations of magnetization in magnetic crystals, deduced similarly; no alternative technique is available.
(v) Energy of thermally excited molecular oscillations and rotations in crystals, liquids or gases—analogous to infrared spectroscopy but frequently specific to proton motions and applicable to symmetrical molecules which are otherwise inaccessible.
(vi) Static pair and triplet distributions of atoms in a liquid (including the partial distribution functions in a binary system).
(vii) Time-dependent pair distribution in a liquid, which is related to the cooperative modes of motion.
(viii) Self diffusion of atoms on atomic distance and time-scales (related to small angle incoherent scattering).
(ix) Time-dependent correlations of magnetization near and above the magnetic critical point.
(x) States of electrons in conducting substances (related to the magnetic scattering).
(xi) Distribution of atomic or magnetic distortion around atomic scale defects in crystals has been measured.
(xii) Dynamics of different molecular groups in one molecule has been measured by the proton/deuteron substitution technique.

Atomic Distributions in Crystals

The determination of the positions of light atoms in structures and the differentiation of atoms of similar atomic number are possible with neutrons. This has removed serious restrictions on X-ray work which have held up studies of hydrogen bonding, hindered rotations of ions, hydride structures, heavy element oxides, carbides and nitrides, and studies of ordering in many interesting metallic systems.

Samples with linear dimensions of about 1 mm are used and through the use of automatic diffractometers the reciprocal lattice can be explored in three dimensions in a reasonable time. Recent work has not only resulted in a greatly increased accuracy of parameter determination (for example, a standard deviation of ±0·008 Å is claimed for the location of the hydrogen atoms in hexamethylene tetramine) but has provided general conclusions on groups and series of compounds, simply because of the greatly increased output of information. For example, the high accuracy of hydrogen location which neutron beams permit has made it possible to examine accurately the problem of random and rigid body motion in relatively simple molecules which can be studied theoretically by quantum chemists. Thus naphthalene, resorcinol, salicylic acid and hexamethylene tetramine have been studied in detail by university groups at Harwell. In the case of hexamethylene tetramine it has also been demonstrated that a concurrent refinement of X-ray and neutron data produces a more satisfactory result than the use of either set of data separately.

Magnetization Distributions and Liquid Structure

Although the close connexion between strong magnetic properties and the anti-symmetry condition for electron wave functions was recognized by Heisenberg in 1926, it is still impossible to predict the magnetic structures of matter. Even for technologically important materials, such as ferrites and garnets, there is only a skeleton empirical knowledge of the facts, provided mostly by neutron techniques. The magnetic electrons are the outer electrons and are interesting for chemical as well as physical reasons.

Scattering of polarized neutrons from unsymmetrical antiferromagnets, for example, MnF_2, shows the transfer or sharing of spin between "magnetic" ions (M_n^{++}) and "non-magnetic" ions (F^-). This is the most direct evidence of covalency available and is in a form suitable for direct comparison with theory.

The detailed understanding of the liquid state remains one of the outstanding problems of condensed matter physics. Measurements of the liquid structure factor for monatomic liquids can be made to higher accuracy than with X-rays and good data can be obtained with the samples inside a cryostat, furnace or pressure containers. This makes it possible to measure pressure and temperature derivatives which, in addition to providing data on the pair distribution function, allows the triplet and higher order distributions to be studied. Thus a new insight into liquid structure is being achieved.

Of special interest are studies on binary (*A–B*) systems (for example, alloys or fused salts) where by isotopic substitution three or four separate diffraction patterns can be obtained for a single system. From these data the three pair distribution functions (*AA*, *BB* and *AB*) can be obtained and which transform treatment of binary systems from "primitive guesswork" into a "scientific subject". Examples are provided by the resistivity of a binary alloy (CuSu) and the interaction potential in a fused salt (CuCl).

Crystal Lattice Vibrations

Neutrons are unique in that their energies are thermal when their wavelengths are atomic. This makes possible precisely defined experiments on phonons and spin waves, which have led to a new understanding of interatomic forces.

The first vibrational studies to be completed in detail (at Chalk River in Canada) showed that although some of the forces between atoms in a metal were best thought of as comparatively local, some are of very long range, and probably represent forces derived from the electron gas in some way. In ionic and covalent crystals the phonon frequencies showed some unpredicted effects, now regarded as evidence for polarization by deformation. A study of lead has shown anomalies which are due to the interaction between the electron Fermi surface and the phonons—the interaction which is the basis of the current explanation of superconductivity. Solid helium has been investigated in a range of temperature and pressure and the phonon dispersion relations for argon at 4 K have been determined.

Inelastic scattering of neutrons also provides a means of studying directly fluctuations in the magnetization of crystals (spin waves), for example, in ferromagnets, antiferromagnets and ferrimagnets. Again this is the only available method for work on bulk substances (there is a resonance technique available for thin films of ferromagnets). In metallic systems the inadequacies in the original theoretical formulation of the problem are now clearly exhibited for the first time.

Liquid Dynamics

Inelastic scattering from liquids facilitates study of the dynamics of atoms or molecules. In the study of diffusion it is possible to examine the details of a single diffusive step on a time scale of about 10^{-12} s. The scattered neutron spectrum can be compared with the predictions of diffusion theories, and it can be shown, for example, whether the diffusion process consists of atoms jumping from site to site, as in a solid, or of combined movements of groups of atoms, analogous to Brownian motion, or, as actually occurs, the process is a combination of several mechanisms. The significance of this knowledge in the study of chemical reactions has been realized, but the accuracy of individual experiments, and the throughput of different compounds, is severely limited by present fluxes and facilities.

The discussion of the short wavelength collective motion in liquids is complicated because these modes are heavily damped due to their strong interaction with one another. Their behaviour is described by current–current correlation functions which can be measured only by the neutron scattering technique. This area is stimulating theoretical work on many body systems, and this in turn leads to more exacting neutron inelastic scattering studies of simple liquids. Such work is relevant also to the study of the dynamics of amorphous materials. Here the problems are usually greater, for molecular materials are being examined, and work so far (on SiO_2, for example) has produced results on a limited number of modes only.

Molecular Dynamics

A major growth point has been the application of neutron techniques to the study of molecular spectroscopy in gases, liquids and solids of interest to chemists. The studies include work on small molecules, on synthetic and natural macromolecules and on molecules absorbed on surfaces, and they complement infrared and Raman spectroscopic techniques. The special features of the neutron technique include the removal of the restrictions imposed by optical selection rules and very different (often simpler) intensity relationships. The intensity of a vibrational band is simply related to the amplitudes of nuclear displacement in the normal mode. Particular use is made of the abnormally high scattering cross-section of the proton; substitution of ^2H for ^1H greatly reduces the scattering cross-section and provides an important diagnostic tool. Recent applications include studies of (i) torsional modes in $CH_3{\cdot}CH_3$, $CH_3{\cdot}CF_3$ and $CH_3{\cdot}CCl_3$, (ii) the dynamics of polymer main-chains, side-chains and crystal lattices to obtain information relevant to structure–property relations in polymers, and (iii) direct observation of hydrogen bonds.

Defect and Domain Scattering

Long wavelength neutrons can be scattered from crystals only if deviations from the crystal perfection are present, so that the scattering of neutrons of 4–10 Å gives a powerful method of studying defects on a size scale of 2–20 Å. This is just below the resolution of the electron microscope and there is considerable promise in this method for the study of crystal damage caused by, for example, radiation.

An important recent development is the investigation of diffuse scattering by non-stoichiometric compounds and solid solutions. It is already clear that the technique has great potential for providing information about clustering and short range ordering of atoms in various solids.

High Flux Reactor Proposal

The justification of a large neutron beam facility lies in the overall impact made by neutron experiments on the study of condensed matter. Generally a neutron experiment is so definitive that it is used as the basic experiment in a field, and the interpretation of the data obtained by other methods is greatly improved through the use of reliable molecular knowledge. Thus neutron scattering has a central role, well illustrated by the examples of magnetic structure and crystal lattice dynamics given here. In these cases the entire field rests on the neutron data, which are used by theoreticians and experimentalists for further interpretations and extensions of knowledge. Thus the important ideas in a discussion of neutron beam facilities are those involving the broader implications of neutron work, the strengthening or weakening of entire fields of study and the basic significance of the study of condensed matter in the life of mankind.

The gradual development of neutron scattering techniques through the 1950s had led by 1960 to a relatively sophisticated programme for the study of condensed matter. This was recognized by the International Atomic Energy Agency (IAEA) which started in 1960 a series of conferences on the inelastic scattering of neutrons in solids and liquids. Four such conferences have been held. In the United Kingdom the main team doing this work was concentrated at Harwell, but other groups (chiefly at Cambridge) were involved. Thus in 1960 the future heralded the expansion and development of a new and very powerful technique. Many very important problems, concerned with the study of condensed matter and with significant practical applications, were waiting to be investigated, and many scientists both within the Atomic Energy Authority (AEA) and in universities were anxious to help.

But the reactors used until this time had been designed for other purposes—primarily the irradiation of materials of significance to the reactor power programme: the Harwell reactors DIDO and PLUTO are called materials testing reactors for this reason. As a result they had three basic disadvantages for thermal neutron scattering experiments. These concerned the shield, the building and the flux. First, the shield of a materials testing reactor is arranged to be just thick enough to reduce the radiation to a safe biological level. But this level produces a very high background in thermal neutron scattering experiments and a substantially thicker and more efficient shield is required in a purpose built reactor. Second, the building which houses DIDO and PLUTO is too small to contain much of the sophisticated neutron scattering equipment. When a reactor is used to irradiate materials there is little need for horizontal space: most of the specimens are loaded in vertical rigs and headroom above the reactor is the major requirement. Horizontal space is needed only for coffins to accept active materials from the horizontal holes. For this reason the containment buildings of the DIDO and PLUTO reactors are very small, and when the neutron scattering apparatus was installed both the design and operation of the spectrometers were restricted. Third, the flux available from these reactors is not optimized for neutron beam experiments. The fuel elements are spaced at distances which allow samples to be placed between them, and the thermal neutron flux then peaks at the irradiated specimens. Unfortunately, neutron beam tubes are placed at the ouside of the core and are therefore not in the region of maximum flux. In the design of a thermal neutron beam reactor, the fuel elements are placed as close together as possible so that the core has a very high flux of fast neutrons. These fast neutrons enter the heavy water around the core and are thermalized, producing a peak in the thermal flux at approximately the position of the neutron beam tubes. This design advantage together with a higher power level will give the HFBR a factor of 15 advantage in flux over DIDO and PLUTO.

Table 1. RESEARCH REACTOR COMPARISONS

Country	Reactor	Date	Relative flux for typical beam user
UK	DIDO	1956	1
UK	PLUTO	1957	1·5
UK	HERALD	1960	0·3
USA	HFBR	1966	10
USA	HFIR	1967	15
Germany/France	HFBR	1971	20
Holland	HFR	1962	1·5
Belgium	BR2	1961	1·5
Sweden	R2	1960	1·5

During the 1960s, the horizontal holes of DIDO and PLUTO at Harwell and HERALD at Aldermaston were progressively taken over by the neutron beam programme until now they are almost all used for neutron spectrometers. Because of the low neutron fluxes, very substantial efforts were made to maximize the efficiency of the neutron spectrometers and of the detecting and data handling systems. In addition the number of university users increased during this period from six to approximately 100: thus the development anticipated in 1960 took place in spite of the restrictions in the use of the existing reactors. This, of course, demonstrates the attraction of the neutron scattering technique for the study of condensed matter.

A comparison of research reactors available or under construction in various countries is shown in Table 1 (the Germany/France figure is an estimate). It shows that the DIDO and PLUTO reactors are old compared with the reactors in other countries and the newer reactors in the USA and Europe have a significant flux advantage (as well as more modern experimental facilities). Until 1960 the reactors at Harwell had kept in step with developments in other countries, but we have now fallen materially behind.

It was in anticipation of these developments that a group of reactor designers and neutron beam experimentalists proposed in 1960 that a new HFBR be built at Harwell. A design study was made of this project and in 1961 a proposal was ready.

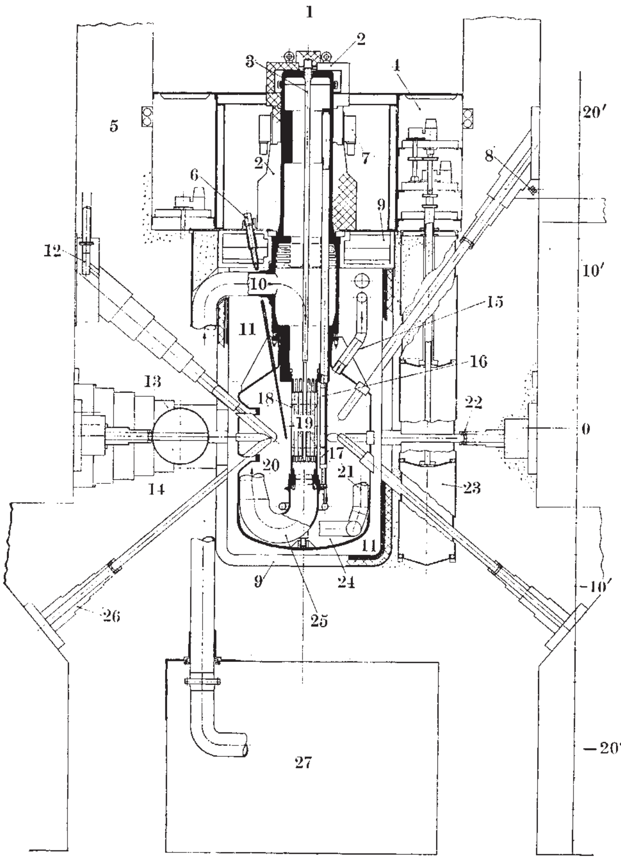

Fig. 1. Cross-section of the proposed high flux beam reactor. 1, Fuel element handling room; 2, shielding; 3, in-core rig; 4, beam shutter drives; 5, high density concrete biological shielding; 6, irradiation facility; 7, control rod drives; 8, removable shield block; 9, thermal shield; 10, core D_2O inlet; 11, CO_2 void; 12, cold source services; 13, removable shield; 14, cartwheel beam shutter; 15, reflector inlet; 16, upper control rods; 17, lower control rods; 18, zirc II pressure barrel; 19, core; 20, core D_2O outlet; 21, reflector D_2O outlet; 22, aluminium windows; 23, split rotating beam-shutter; 24, reflector tank–D_2O at atmosphere pressure; 25, core circuit–D_2O at 400 pounds/inch^2; 26, low angled beam hole; 27, sub-pile D_2O plant room.

This design was discussed later as the possible basis for a European international project, but no agreement was reached on various aspects of siting and finance, so that the project was shelved. In 1967 the French and German Governments set up a bilateral agreement to construct such a reactor at Grenoble, and this is now being built. The general outline design for a reactor suitable for the range of experiments appropriate to the United Kingdom has been completed, and there is strong user support for its construction. The proper channels of funding such a project of both technological and scientific promise involve more than one government department. The considerable expenditure involved (the Franco-German project budget is £11 million), naturally comes under close scrutiny when the national scientific budget is under great pressure, and the matter is still *sub judice*.

Matters singled out for careful study include the relative merits of a continuous reactor and a pulsed neutron source. The comparison is complicated because the continuous source techniques are better developed than the pulsed source techniques, and the two techniques are not equally applicable to all classes of experiments. The continuous beam reactor is clearly superior in experiments which require a continuous neutron beam and experiments based on long wavelength neutrons (greater than 4 Å). On the other hand, for some neutron time of flight experiments (for example, with relaxed collimation and for short neutron wavelengths) the pulsed neutron source will be superior. In the former category are most of the experiments with crystals, including the study of structure and dynamics using both the nuclear and the magnetic scattering processes. The long wavelength field includes studies of defects and many transient phenomena in liquids and polymers on a long time scale. On the other hand, in the category of time of flight experiments (with $\lambda < 3$ Å), concern is chiefly with inelastic scattering processes on a short time scale, particularly for the study of liquids, polymers and molecular spectroscopy. Experiments of this type are less detailed in interpretation at present, though they include significant work in a rapidly developing scientific area. Thus the decision between these sources rested on the balance of user interest. Because time of flight experiments of high quality can be done on the reactor, the larger number of users in the former category persuaded the minority in the latter category to accept the HFBR as the preferred source. This choice also satisfies the requirement that it is possible to prosecute a balanced programme on any major neutron source.

The HFBR project is ten years old and the present design is not far removed from the design of 1960, so that it might be asked whether the reactor is obsolete ? There are several reasons why this is not so. First, the reactor field is reaching a plateau in its development determined by the heat removal rates from the fuel elements (which in the HFBR are approximately 1 kW/5 cm of fuel element or 100 MW in $0{\cdot}16$ m^3). It is difficult to increase the heat rating and hence the flux without making major, and very expensive, changes in the design amounting to the development of a new reactor system. These changes might include very high flow rates of cooling water through the use of very high pressures or might involve the use of a liquid fuel rather than a standard metallic fuel element, but do not offer obvious advantage at a reasonable cost. Nevertheless, the 1970 design does include improvements in the design of the reactor components which have occurred during the intervening ten years. Second, a more significant factor is the balance between the different types of user or, in other words, the overall scientific programme. Special Science Research Council (SRC) committees have confirmed that the outcome of the discussions on the type of source to be preferred, held during the 1960s, is still valid, and therefore the basic decision on the type of source is still valid.

SRC Neutron Beam Programme

The SRC makes use of neutron beam time on three reactors owned by the AEA: these are the HERALD reactor at Aldermaston and the DIDO and PLUTO reactors at Harwell. The flux available at the beam holes of HERALD is about one-third of that available at the DIDO and PLUTO reactors, but because many users cannot be accommodated on the Harwell reactors it is necessary to use this reactor as well. This programme is managed by the SRC Neutron Beam Committee which reports annually on progress. Its scientific reports cover the fields in which predictions of neutron beam interest have been made during the past few years and university scientists are translating into successful experiments many of the approaches which have been claimed to be practicable.

The value of neutrons to chemists as well as physicists is clearly evident in the scope of the reports on chemistry submitted by the Neutron Beam Committee. Approximately half of the scientists using the neutron beam facilities are from university chemistry departments. Some of the areas they cover were listed earlier in this article. Some proposals have been made and initial experiments conducted on biological problems with neutrons, but this field is in its infancy. It is anticipated, however, that it would expand rapidly through the improvements provided by the HFBR. An analysis of the present programme into categories of use is given in Table 2.

Table 2. DISTRIBUTION OF UNIVERSITY USERS OF AEA REACTORS

Field	No. of users
Chemical crystallography	30
Magnetic crystallography	18
Inelastic excitation of solids	21
Liquids	11
Defects in solids	13
Properties of neutrons	2
Total	95

Each physical technique seems to follow a general pattern of development. First, it is developed by the physicists concerned; in this case the nuclear physicists. It is then taken over by other physicists for use in basic research; in this case the condensed matter physicists. Subsequently other scientists become interested (in this case chemists and biologists) and use the technique. If as a result the technique is shown to have a wide scientific application it is used increasingly for applied problems in many industries. This pattern of development can be seen, for example, in X-rays, in infrared absorption, nuclear magnetic resonance and so on.

It seems likely that neutron scattering is following this general pattern, and is entering the final phase of its application to applied and industrial problems. The number of industrial scientists using the neutron spectrometers at Harwell and Aldermaston is small but increasing: their scientific and technical interest covers the same general field as the SRC scientists, though the motivation of their companies is industrial development. Although their work includes the structure and the dynamics of condensed matter, in many cases the samples are complex. This is partly because they are a mixture of many species and partly because large molecules are involved. Thus as the neutron scattering technique is developed for chemical and biological purposes we may expect parallel development in its use for applied and industrial applications. In some areas this is aided by the high efficiency of modern neutron spectrometers and the fact that they operate continuously and on a routine basis.

Present Proposal

Fig. 1 shows the cross-section of the proposed HFBR. Twelve horizontal beam holes are fed by different neutron sources. Several observe the ambient neutron spectrum in the heavy water while others are connected to the cold neutron source for long wavelength experiments and a group is connected to the hot neutron source for short wavelength experiments. In addition to these horizontal holes several inclined holes (both upwards and downwards) have been included. The flux viewed by these holes is lower than that seen by the horizontal holes but is still higher than that available on existing reactors. Also shown in the figure are the multi-hole arrangements for the cold and hot neutron sources. A limited number of irradiation facilities are provided also. It is proposed to operate the reactor at 100 MW and the maximum flux available would be $1\cdot5\times10^{15}$ neutrons/cm^2 s. The fuel is an Al/U alloy cooled by heavy water under a pressure of $28\cdot2\times10^4$ kg/m^2.

It is anticipated that after the project has been approved it will take five years to reactor criticality, and all twenty or so experiments should be in full swing after two more years. Because the neutron spectrometers could be constructed while the reactor is being built the experimental programme could commence as soon as the reactor is operating continuously at full power. At present it is proposed that the new spectrometers should include all the major types in use at Harwell: but the position will be reviewed at stages so that new techniques can be included in the instruments for the HFBR. Present proposals include the provision of several time of flight spectrometers, several three-axis neutron spectrometers as well as diffractometers for single crystal analysis. Other specialized instruments for long wavelength work and defect studies are proposed, making in all twenty-seven different spectrometers.

The higher flux may be used in various ways, for example; smaller specimens may be examined, higher resolution in energy and/or momentum may be used, the wavelength range may be extended to higher or lower limits, the statistical accuracy of the data may be improved, the study of weak scattering processes in the presence of strong ones may be improved and so on. The choice between these alternatives is open to the scientist concerned, for neutron spectrometers can be designed with flexibility in mind. Once again the neutron beam work is characterized by its breadth, and when transferring to a new reactor the neutron beam scientist reconsiders his experimental programme and makes use of the higher flux in one (or more) of the six or so ways open to him.

With a reactor of the type proposed the university teams would be able to consolidate their neutron beam programmes and to maintain the lead the United Kingdom has given in the field of condensed matter science.

I thank Dr W. M. Lomer for helpful suggestions and Professor E. W. J. Mitchell for permission to use some material from his SRC report on the scientific case for the HFBR.

[1] *Thermal Neutron Diffraction* (edit. by Willis, B. T. M.) (Oxford University Press, 1970).
[2] *Inelastic Scattering of Neutrons* (IAEA, Vienna, 1969).